Interfaces and mixing: Nonequilibrium transport across the scales

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Interfacial transport and mixing are nonequilibrium processes coupling kinetic to meso- and macroscopic dynamics. These processes play an essential role in fluids, plasmas, and materials, from celestial to atomic events. Addressing the societal challenges posed by alternative energy sources, efficient use of nonrenewable resources, and purification of water requires an improved understanding of the nonequilibrium dynamics, interfacial transport, and mixing. This special feature issue builds upon recent achievements in understanding interfacial transport and mixing using theoretical analysis, large-scale numerical simulations, laboratory experiments, and technology developments. It brings together works in fluid dynamics, plasmas, applied mathematics, chemistry, material science, geophysics, and astrophysics. This collection of papers explores the state of the art in areas of interfaces and nonequilibrium transport and suggests future research directions in the field.

This special feature issue of PNAS presents a number of outstanding contributions in "Interfaces and mixing: Nonequilibrium transport across the scales." This issue is associated with the symposium on "Interfaces and Mixing" invited by the US National Academy of Sciences in 2017 and is a part of the program "Turbulent Mixing and Beyond" founded in 2007 with the support of the international scientific community and national and international funding agencies and institutions (1–6). This issue, the symposium, and the program bring together researchers from many areas of science, mathematics, and engineering and focus their attention on fundamental problems of interfaces, mixing, and nonequilibrium dynamics (1–6).

Interfaces, mixing, and nonequilibrium dynamics govern a broad range of phenomena in nature and technology, in high- and low-energy density regimes, at astrophysical and at atomic scales (1–19).

Examples include supernovas, molecular hydrogen clouds, and accretion disks; magnetic, magneto-inertial, and inertial confinement fusion; light–materials interaction, impact-induced materials transformations, and materials melting and evaporation; strong shocks, explosions, and detonations; dynamics of supercritical, reactive, and chemistry-driven fluids; convection in stellar and planetary interiors and mantle–lithosphere tectonics; flows in ocean and atmosphere; and environmental flows (1–19). A good grip on interfaces and mixing and their nonequilibrium dynamics is crucial for cutting-edge technology in nanofabrication and free-space optical telecommunications, in efficient use of nonrenewable resources, and in purification of water, as well as in traditional industrial applications in the areas of aeronautics and aerodynamics (1–19). In some of these applications (e.g., combustion processes), interfacial mixing should be enhanced; in some others (e.g., inertial confinement fusion) it should be mitigated and tightly controlled (1–19). However, in all these circumstances, we have to understand the fundamentals of interfacial mixing and nonequilibrium dynamics, be able to gather high-quality data while deriving knowledge from these data, and, ultimately, achieve better control of these processes, from atomic to macroscopic scales.

The field of interfaces, mixing, and nonequilibrium dynamics provides a source of paradigm problems in physics, mathematics, and engineering (1–3). They span areas of theoretical physics and applied mathematics, materials science and chemistry, fluids and plasmas, astrophysics and earth sciences, probability and statistics, data processing and computations, and many other areas (1–19).

Interfacial transport and mixing couple kinetics from atomic to continuous scales and are exceedingly challenging to study (1–19). Their dynamics involve sharply and rapidly changing flow fields and may also include strong accelerations and shocks, radiation transport and chemical reactions, and transport of species and electric charges, among other effects (1–19). At microscopic atomic scales strong energy fluctuations and complex electron transfer processes induce a broad range of interactions, timescales, chemical reactions, products, and complexes, far from thermal equilibrium. At mesoscopic kinetics scales, nonequilibrium processes depart dramatically from standard

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scenarios of Gibbs ensemble averages and the quasi-static Boltzmann equation (1–19). At macroscopic continuous scales, with flows fields and phases formed, interfacial mixing and nonequilibrium dynamics are nonlocal, inhomogeneous, anisotropic, and statistically unsteady; their invariance, correlations, and spectra differ substantially from those of canonical turbulence (1–19).

The theoretical description of interfacial mixing and nonequilibrium processes is intellectually challenging, since it has to account for the multiscale, multiphase, nonlinear, nonlocal, and statistically unsteady character of the dynamics and accurately solve the singular boundary value problem and the ill-posed initial value problem (1–19). Their numerical modeling perpetually broadens horizons of massive computations while demanding improvements in numerical methods to track interfaces, capture sharp and rapid changes of flow fields, and account for the scale coupling over large spans of temporal and spatial scales (1–19). On the experimental side, interfacial mixing and nonequilibrium processes are a challenge to implement and study systematically in a well-controlled laboratory environment (1–19). This is because they are transient and sensitive to deterministic conditions, and their dynamics impose tight requirements on the accuracy and resolution of measurements and on data acquisition rates (1–19). Furthermore, a systematic interpretation of these processes from the data is neither easy nor straightforward, since the processes are statistically unsteady and impose as a primary concern the influence of an observer on observational results (1–19). At the same time, interfacial mixing and nonequilibrium processes may lead to self-organization and order and may thus expand opportunities for diagnostics and control of nonequilibrium dynamics in nature and technology, for better understanding fundamentals of interfacial mixing, and for capturing the interplay of particles and fields (1–19).

This issue reports significant successes that have been achieved recently in understanding interfaces, mixing, and nonequilibrium dynamics based on theoretical analysis, large-scale numerical simulations, laboratory experiments, and technology development (20–34). These include theoretical approaches for handling complex multiscale, nonlocal, and statistically unsteady dynamics and boundary value problems; developments of efficient Eulerian and Lagrangian methods of large-scale numerical modeling; advancements in laboratory experiments in low- and high-energy density regimes; and possibilities for dramatic improvements in precision, accuracy, dynamic range, reproducibility, and data acquisition rate with the use of modern technologies (20–34).

These successes as well as the striking similarity in behavior of interfacial mixing and nonequilibrium dynamics in the vastly different regimes make this moment right to integrate our knowledge of the subject and to further enrich its development (1–6, 20–34). We can explore properties of interfacial mixing, study fundamentals of nonequilibrium dynamics, touch the matter at extremes, and develop a unified description of particles and fields on the basis of synergy of theory, simulations, and experiment (1–6, 20–23). At this right moment, we can apply the fundamental knowledge of interfacial mixing and nonequilibrium dynamics for addressing contemporary challenges of science, technology, and society.

Hence with the support of the National Academy of Sciences, the international scientific community, and national and international institutions, we introduce this special feature issue on "Interfaces and Mixing" in PNAS (1–6, 20–34). The contributing authors are both senior and junior researchers from universities and national laboratories and from national and international communities, including members of the National Academies of Sciences in the United States and from abroad and graduate and undergraduate students.

The papers in this issue focus on fundamentals of interfacial mixing and nonequilibrium dynamics, from particles to fields, across the scales and disciplines. These works represent frontier research in interaction of molecules and chemical reactions, interfacial dynamics and nonequilibrium processes, fluid turbulence and turbulent mixing, supernovas and nuclear synthesis, formation of fluid phases at molecular scales, first-principles–based reaction kinetics, ion transport of nanoscales, electric and magnetic fields structures, dynamics of high-energy density plasmas, subdiffusive and superdiffusive transport, formation of vortices in geophysical flows, diffusiophoresis of charged particles, and electron transport in macromolecules. They motivate the discussions of rigorous mathematical problems, theoretical approaches, and state-of-the-art numerical simulations along with advanced experimental methods and technological applications (20–32).

The contributions in this issue include 6 perspective papers and 7 research papers (20–32). They are ordered from perspective to research papers, from macroscopic to microscopic scales, and from fundamentals to applications. Refs. 20, 25, and 29 (with invited authors Sreenivasan, Abarzhi, and Nepomnyashchy, respectively) explore the fundamentals of turbulent mixing and interface dynamics at macroscopic scales. Refs. 22–24 and 26 (with invited authors Goddard, Ilyin, Anisimov, and Schlossman, respectively) investigate nonequilibrium dynamics at microscopic scales. Refs. 21, 27, and 28 (with invited authors Arnett, Remington, and Gekelman, respectively) analyze complex processes in high- and low-energy density regimes. Refs. 30–32 (with invited authors Haller, Prieve, and Kais, respectively) examine applications of multiscale dynamics in nature and technology.

The perspective paper by Sreenivasan (20) investigates the turbulent mixing paradigm—the mixing and advection of distinct substances by a turbulent flow. The case of passive scalar mixing is of particular interest—when the substance is characterized by its concentration so that the mixing does not influence the flow itself. The author discusses how a turbulently mixed state depends on the flow viscosity and the scalar diffusivity, identifies the fundamental aspects of turbulent mixing, and summarizes ideas that explore the universal and anomalous scaling properties of turbulent mixing and help us to appreciate its physics in diverse circumstances.

The perspective paper (21) explores the supernova, nuclear synthesis, fluids instabilities, and interfacial mixing. Supernovas and their remnants are a central problem in astrophysics due to their role in the stellar evolution and nuclear synthesis. A supernova's explosion is driven by a blast wave causing the development of Rayleigh–Taylor instabilities and leading to the intensive interfacial mixing of the star's materials. The authors apply group theory to analyze properties of Rayleigh–Taylor dynamics, discover the subdiffusive character of the blast wave-induced interfacial mixing, and reveal the mechanism of energy transport enabling nuclear synthesis in supernovas.

The perspective paper by Goddard and coworkers (22) focuses on developments of rigorous yet practical methods that extend the scale of the atomistic simulation by several orders of magnitude to describe realistic chemical processes while retaining quantum mechanics accuracy. These developments can enable continuum modeling of interfacial transport to incorporate the relevant chemistry. The work is particularly focused on the recent progress in accomplishing these extensions in first-principles–based atomistic simulations and on the strategies being pursued to increase the accuracy of very large scales while dramatically decreasing the computational effort.

The perspective paper by Ilyin et al. (23) presents the use of in silico approaches in extracting the reaction mechanisms and kinetic parameters describing complex chemical processes in reactive systems. The authors develop a reactive molecular dynamics to kinetics method for direct analysis of reactive systems with no preconceived ideas about the chemistry. They find that even simple chemical reactions can be accompanied by complex processes with transient states and with energy fluctuations that may lead to chemistry-induced instabilities.

The perspective paper by Zhakhovsky et al. (24) links nonequilibrium dynamics of particles and fields by considering phase transitions associated with processes of evaporation and condensation. The authors apply the Boltzmann kinetic equation and the molecular dynamics simulations to capture the strongly coupled mass and heat transports. This strategy bridges the gap between the atomistic representation of the complex motion of atoms and probabilistic evolution of distributions of vector and scalar fields in conditions far from equilibrium.

The perspective paper by Abarzhi et al. (25) focuses on the classical problem of stability of a phase boundary—a fluid interface with mass flow across it. This problem is even more challenging than that of the Clay Institute Millennium problem on the Navier–Stokes equation and turbulence. The work develops the general theoretical framework directly linking the microscopic interfacial transport to macroscopic flow fields, discovers the inertial mechanism of interface stabilization and the fluid instability, which has not been discussed before, and identifies the destabilizing effect of energy fluctuations (e.g., chemical reaction). Some of the applications for this theory include fusion, supernovas, fossil fuel extraction, and nanofabrication.

The research paper by Liang et al. (26) studies the transport of metal ions across the liquid–liquid interface between an aqueous ionic solution and an organic solvent. To probe fast molecular processes on nanometer-length scales in the presence of interfaces, the authors apply state-of-the-art techniques to characterize these processes using X-ray surface scattering of intermediate states and observe directly the essentially nondiffusive transport of ions across the interface.

The research paper by Remington et al. (27) reviews experiments on Rayleigh–Taylor instabilities in high-energy density settings and explores the properties of matter at extreme conditions. The experiments report observations of a more ordered character in Rayleigh–Taylor dynamics in 3 regimes—at ablation fronts, behind radiative shocks, and due to material strength. These unique experimental data may help to better understand a broad range of processes in high-energy density plasmas.

The research paper by Gekelman et al. (28) analyzes magnetic flux ropes in plasma experiments. Such structures are commonly observed in the corona of the Sun, the Earth's magneto-tails, and near Mars and Venus. The unique laboratory environment enables the detailed study of interactions between multiple ropes and their topological properties such as helicity and writhing. The authors report on spiky potential and magnetic fields associated with the ropes and show that the structures are essentially chaotic.

The research paper by Zaks and Nepomnyashchy (29) contributes to studies of the Navier–Stokes equation by investigating the transport of passive tracers in steady laminar plane flows through obstacles—arrays of solid bodies or steady vortices. The authors find that although the transport is deterministic, it can be anomalous, i.e., be subdiffusive or superdiffusive depending on the flow conditions, due to singularities of the passage times near the obstacles and decorrelations.

The research paper by Beron-Veraa et al. (30) presents the multiscale data analysis of geophysical flows—the circulation in the Gulf of Mexico. To analyze satellite-based data of the ocean flows, the authors apply formal nonlinear dynamics tools and illustrate the emergence of coherent Lagrangian swirls among the submesoscale motions in the ocean flows. This approach permits more realistic modeling of geostrophic multiscale flows and climate change.

The research paper by Prieve et al. (31) describes the complexity of diffusion processes in realistic environments. These authors study experimentally the diffusiophoresis—the process of the migration of a colloidal particle through a viscous fluid—at extreme conditions of high salinity. They find that diffusiophoretic speeds are measurable even for concentrations approaching the solubility limit and discuss industrial applications of their results.

The research paper by Yeh et al. (32) presents the complexity of nonequilibrium dynamics in macromolecules. The authors investigate the long-lived coherences observed in natural and artificial light-harvesting systems, by attributing them to the mixing of electronic and vibrational degrees of freedom of pigments. The authors elucidate the relationship between the coherence lifetime, the electronic energy fluctuation, and the vibrational relaxation dephasing pathways.

This special feature issue explores and assesses the state of the art in interfaces, mixing, and nonequilibrium dynamics and charts directions of future research in this field. We hope that the world of interfacial mixing and nonequilibrium dynamics will be exposed to the international scientific community and that the inquisitive minds will be captured and fascinated by the opportunity to elaborate the concepts, whose lucidity and simplicity can cut through the complexity of the problem.

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